

**TRANSMITTAL LETTER TO THE  
UNITED STATES RECEIVING OFFICE**

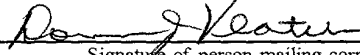
Date	25 February 2002
International Application No.	PCT/EP00/08070
Attorney Docket No.	Mo6979/LeA 33,536

**I. Certification under 37 CFR 1.10 (if applicable)**

ET671453725US
Express Mail mailing number

25 February 2002
Date of Deposit

I hereby certify that the application/correspondence attached hereto is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to Assistant Commissioner for Patents, Washington, D.C. 20231.


Signature of person mailing correspondence

Donna J. Veatch
Typed or printed name of person mailing correspondence

**II. ☒ New International Application**

TITLE	Multilayer Platic Composites and a Method for the Production Thereof
-------	--

Earliest priority date (Day/Month/Year)
30 Aug 1999 (30.08.99)

**SCREENING DISCLOSURE, INFORMATION:** In order to assist in screening the accompanying international application for purposes of determining whether a license for foreign transmittal should and could be granted and for other purposes, the following information is supplied. (Note: check as many boxes as apply):

- A. ☒ The invention disclosed was **not** made in the United States.
- B. ☒ There is no prior U.S. application relating to this invention.
- C. ☐ The following prior U.S. application(s) contain subject matter which is related to the invention disclosed in the attached international application. (NOTE: priority to these applications may or may not be claimed on form PCT/RO/101 (Request) and this listing does not constitute a claim for priority.)

application no.		filed on	
application no.		filed on	

- D. ☐ The present international application contains additional subject matter not found in the prior U.S. application(s) identified in paragraph C. above. The additional subject matter is found on pages  and ☐ DOES NOT ALTER ☐ MIGHT BE CONSIDERED TO ALTER the general nature of the invention in a manner which would require the U.S. application to have been made available for inspection by the appropriate defense agencies under 35 U.S.C. 181 and 37 CFR 5.1. See 37 CFR 5.15

**III. ☐ A Response to an Invitation from the RO/US. The following document(s) is(are) enclosed:**

- A. ☐ A Request for An Extension of Time to File a Response
- B. ☐ A Power of Attorney (General or Regular)
- C. ☐ Replacement pages:

pages		of the request (PCT/RO/101)	pages		of the figures
pages		of the description	pages		of the abstract
pages		of the claims			

- D. ☐ Submission of Priority Documents

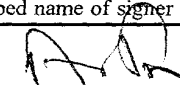
Priority document		Priority document	
-------------------	--	-------------------	--

- E. ☐ Fees as specified on attached Fee Calculation sheet form PCT/RO/101 annex

**IV. ☐ A Request for Rectification under PCT 91 ☐ A Petition ☐ A Sequence Listing Diskette**

- V. ☒ Other (please specify): Preliminary Amendment; Abstract page  
PTO form 1449  
Drawing

The person  
signing this  
form is the:

<input type="checkbox"/> Applicant	Aron Preis
<input checked="" type="checkbox"/> Attorney/Agent (Reg. No.) 29,426	Typed name of signer
<input type="checkbox"/> Common Representative	 Signature

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

Mo6979/LeA 33,536

U.S. APPLICATION NO. (If known, see 37 CFR 1.5

To be Assigned 10/088147

INTERNATIONAL APPLICATION NO.

PCT/EP00/08070

INTERNATIONAL FILING DATE

18 August 2000 (18.08.00)

PRIORITY DATE CLAIMED

30 August 1999 (30.08.99)

## TITLE OF INVENTION

Multilayer Plastic Composites and a Method for the Production thereof

APPLICANT(S) FOR DO/EO/US ROEHNER, Juergen; ZANDER, Klaus; GORNY, Ruediger and ROELOFS, Marco

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☐ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☒ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☒ is attached hereto.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:

Abstract page

PTO 1449

Drawing

U.S. APPLICATION NO. (if known) See 37 CFR 1.101

INTERNATIONAL APPLICATION NO.

To be Assessed

10/088147

PCT/EP00/08070

ATTORNEY'S DOCKET NUMBER

Mo6979/LeA 33,536

21. ☒ The following fees are submitted:**BASIC NATIONAL FEE (37 CFR 1.492 (a) (1)-(5)):**

Neither international preliminary examination fee (37 CFR 1.482)  
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO  
and International Search Report not prepared by the EPO or JPO . . . . . **\$1040.00**

International preliminary examination fee (37 CFR 1.482) not paid to  
USPTO but International Search Report prepared by the EPO or JPO . . . . . **\$890.00**

International preliminary examination fee (37 CFR 1.482) not paid to USPTO  
but international search fee (37 CFR 1.445(a)(2)) paid to USPTO . . . . . **\$740.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO  
but all claims did not satisfy provisions of PCT Article 33(I)-(4) . . . . . **\$710.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO  
and all claims satisfied provisions of PCT Article 33(I)-(4) . . . . . **\$100.00**

**ENTER APPROPRIATE BASIC FEE AMOUNT =****CALCULATIONS PTO USE ONLY**

\$ 890.00

Surcharge of **\$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	5 - 20 =	0	x <b>\$18.00</b>

\$

Independent claims	1 - 3 =	0	x <b>\$84.00</b>
--------------------	---------	---	------------------

\$

MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ <b>\$280.00</b>
---	--	--	-------------------

\$

**TOTAL OF ABOVE CALCULATIONS =**

\$ 890.00

☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above  
are reduced by 1/2. +

\$

**SUBTOTAL =**

\$

Processing fee of **\$130.00** for furnishing the English translation later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$

**TOTAL NATIONAL FEE =**

\$ 890.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be  
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **\$40.00** per property +

\$

40.00

**TOTAL FEES ENCLOSED =**

\$ 930.00

**Amount to be  
refunded:**

\$

**charged:**

\$

- a. ☐ A check in the amount of \$ \_\_\_\_\_ to cover the above fees is enclosed.
- b. ☒ Please charge my Deposit Account No. 13-3848 in the amount of \$ 930.00 to cover the above fees.  
A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
overpayment to Deposit Account No. 13-3848 A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card  
information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR  
1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



00157

PATENT TRADEMARK OFFICE

SIGNATURE

Aron Preis

NAME

29,426

REGISTRATION NUMBER

PATENT APPLICATION  
Mo6979  
LeA 33,536

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN APPLICATION OF )  
JÜRGEN RÖHNER ET AL ) PCT/EP00/08070  
SERIAL NO.: TO BE ASSIGNED )  
FILED: HEREWITH )  
TITLE: MULTILAYER PLASTIC COMPOSITES )  
AND A METHOD FOR THE )  
PRODUCTION THEREOF )

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents

Washington, D.C. 20231

Sir:

Prior to its examination kindly amend the enclosed translation of the captioned patent application as follows.

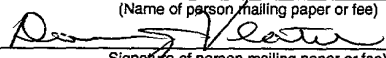
"Express Mail" mailing label number ET671452725US

Date of Deposit February 25, 2002

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Assistant Commissioner of Patents and Trademarks, Washington, D.C. 20231

Donna J. Veatch

(Name of person mailing paper or fee)

  
Signature of person mailing paper or fee)

**IN THE TITLE:**

On Page 1 please amend the title as follows:

--Multilayer Plastic Composites And A Method for the Production Thereof--

**IN THE ABSTRACT:**

Add new page 16, a page containing an abstract that reads as follows:

--Multilayer Plastic Composites And A Method For The Production Thereof

Abstract of the Disclosure

A process for the preparation of a plastic composite is disclosed. The composite contains a first fluoropolymer layer, a second, middle layer of polycarbonate and a third layer of ethylene-vinyl acetate copolymer. The process entails preparing a plastics composite of fluoropolymer and polycarbonate layers and applying the layer of ethylene-vinyl acetate copolymer to the polycarbonate layer.--

**IN THE CLAIMS:**

Please cancel Claim 4.

Please amend Claim 5 as follows:

5. (Once Amended) A process according to Claim 1 characterised in that the fluoropolymer used is polyvinyl fluoride and the polycarbonate used is polycarbonate based on bisphenol A.

**REMARKS**

The present amendment seeks to place the application in better conformance with U.S. practice. A page containing an Abstract of the Disclosure is enclosed. Entry of the amendment is requested.

Respectfully submitted,

By



Aron Preis  
Attorney for Applicants  
Reg. No. 29,426

Bayer Corporation  
100 Bayer Road  
Pittsburgh, Pennsylvania 15205-9741  
PHONE: (412) 777-8343  
FACSIMILE PHONE NUMBER:  
412-777-8363

s/rmc/ap0051

**VERSION WITH MARKINGS TO SHOW CHANGES MADE**

**IN THE TITLE:**

[Multi-layer plastics composites and a process for the preparation thereof] Multilayer  
Plastic Composites And A Method for the Production Thereof

**IN THE ABSTRACT**

Add new page 16, a page containing an abstract as follows:

--Multilayer Plastic Composites and a Method for the Production Thereof

Abstract of the Disclosure

A process for the preparation of a plastic composite is disclosed. The composite contains a first fluoropolymer layer, a second, middle layer of polycarbonate and a third layer of ethylene-vinyl acetate copolymer. The process entails preparing a plastics composite of fluoropolymer and polycarbonate layers and applying the layer of ethylene-vinyl acetate copolymer to the polycarbonate layer.--

**IN THE CLAIMS:**

Please cancel Claim 4.

Claim 5 has been amended as follows:

5. (Once Amended) A process according to [one of claims 1 to 4,] Claim 1 characterised in that the fluoropolymer used is polyvinyl fluoride and the polycarbonate used is polycarbonate based on bisphenol A.

(Name of person mailing paper or fee)



remain solid. Moreover, the coextrusion of the three polymer layers is mentioned in US-A 4 659 625 as a possible method of preparation. It is expressly pointed out in US-A 4 659 625 that the method of preparation must be selected in accordance with the special properties of the polymers used for the three layers.

5

Coextrusion is a particularly advantageous process for the preparation of multi-layer plastics composites. In coextrusion, the layers of the composite are brought together in a coextrusion block as melt layers and then extruded together through a die. In order to produce sheets or films, a slot die, for example, is used during extrusion.

10

The preparation of plastics composites of several layers, wherein one layer comprises an ethylene-vinyl acetate copolymer and one layer comprises polycarbonate is not possible by coextrusion because sufficiently low-viscosity polycarbonate melts have such a high temperature, typically 280°C to 320°C, that the properties of the ethylene-vinyl acetate copolymers are impaired, e.g., due to crosslinking. A deterioration in the optical properties such as, e.g., discolouration and a deterioration in the mechanical properties such as, e.g., strength and elasticity occurs. Such a deterioration is undesirable in terms of the use of the plastics composites, particularly, e.g., in the use in photovoltaic modules.

15

20

Plastics composites of three layers, wherein the first layer comprises a fluoropolymer (particularly polyvinyl fluoride, hereinafter abbreviated to PVF), the second, middle layer comprises polycarbonate (hereinafter abbreviated to PC) and the third layer comprises ethylene-vinyl acetate copolymer (hereinafter abbreviated to EVA) are required, for example, for the manufacture of photovoltaic modules. Their preparation by coextrusion is not, however, possible for the intended application mentioned, for the reasons already given.

25

10088147-022602

Fluoropolymers and polycarbonate adhere very poorly to one another. For this reason, a coupling agent is normally used between the layer of fluoropolymer and the layer of polycarbonate.

5 The object of the present invention is, therefore, to provide a process for the preparation of plastics composites of three layers wherein the first layer comprises a fluoropolymer, the second, middle layer comprises polycarbonate, and the third layer comprises ethylene-vinyl acetate copolymer, which does not have the disadvantages of the prior art.

10 The object according to the invention is achieved by a process for the preparation of plastics composites of three layers, wherein the first layer comprises a fluoropolymer, the second, middle layer comprises polycarbonate and the third layer comprises ethylene-vinyl acetate copolymer, characterised by the following steps to be executed one after the other:

- 15
- a) Preparation of a plastics composite of two layers, wherein the first layer comprises the fluoropolymer and the second layer comprises the polycarbonate,
  - 20 b) Application of the third layer of ethylene-vinyl acetate copolymer to the polycarbonate layer of the plastics composite of two layers.

The present invention thus provides the process mentioned.

25 The process according to the invention has numerous advantages. The required plastics composites of three layers, wherein the first layer comprises a fluoropolymer, the second, middle layer comprises polycarbonate and the third layer comprises ethylene-vinyl acetate copolymer, can be prepared according to the process of the invention in a

simple and inexpensive manner without the ethylene-vinyl acetate copolymer being damaged by heat, e.g., due to crosslinking.

5 The process is solvent-free and therefore advantageous from an economic and ecological standpoint. The process according to the invention permits the continuous preparation of endless plastics composites and, e.g., during a later manufacture of photovoltaic modules, obviates the separate insertion and polishing of the individual layers in each case (films or sheets).

10 The plastics composites according to the invention have good mechanical properties such as, e.g., high strength, high toughness and high elasticity. They have good optical properties such as, e.g., high transparency and high gloss and low production-related discolouration. They have good resistance to chemicals and environmental influences such as weathering and UV radiation. This is particularly the case when at least one of  
15 the three layers has been provided with further additives, such as, e.g., UV stabilisers, absorbers, heat stabilisers, colorants, catalysts, hydrolysis stabilisers, mineral substances, nanoparticles etc. The fluoropolymer layer (optionally also the PC layer) is preferably provided with a sufficient amount of an adequate UV absorber. Moreover, the PC layer is preferably protected by coextrusion, coating or laminating with a UV  
20 absorber-containing material (for example, 2 to 20 wt.% of UV absorber), whether or not the PVF layer contains UV absorber. The layers of the plastics composite adhere well to one another.

25 The process steps a) and b) of the process according to the invention may be carried out one immediately after the other or with an interval of time between them and/or at a distance from one another, e.g., in order to store the two-layer plastics composite on an interim basis. It is preferable to carry out said steps one immediately after the other. In

this case, a heating or cooling of the two-layer plastics composite may take place between process steps a) and b).

5 A preferred embodiment of the process according to the invention is that in which a coupling agent layer is applied to the polycarbonate layer on the two-layer plastics composite which is prepared in process step a), prior to the application of the third layer in process step b).

Suitable coupling agents are, for example, those based on acrylate.

10 A further preferred embodiment of the process according to the invention is that in which the two-layer plastics composite in process step b) has a temperature at which the ethylene-vinyl acetate copolymer does not undergo thermal damage and at which the layer of ethylene-vinyl acetate copolymer is applied as a film to the two-layer plastics composite.

15 In this case, the two-layer plastics composite in process step b) preferably has a temperature from 20°C to 120°C, particularly preferably a temperature from 50°C to 90°C. The EVA film is added preferably at a temperature from 10°C to 60°C, particularly preferably 20°C to 40°C.

20 A further preferred embodiment of the process according to the invention is that in which the two-layer plastics composite in process step b) has a temperature at which the ethylene-vinyl acetate copolymer does not undergo any thermal damage, e.g., due to crosslinking, and at which the layer of ethylene-vinyl acetate copolymer is applied as a melt to the two-layer plastics composite.

25

In this case, the two-layer plastics composite in process step b) preferably has a temperature from 20°C to 120°C, particularly preferably 50°C to 90°C. The EVA melt preferably has a temperature from 80°C to 150°C, particularly preferably from 90°C to 130°C and is applied to the two-layer plastics composite preferably after extrusion through a slot die.

A further preferred embodiment of the process according to the invention is that in which the two-layer plastics composite in process step a) is applied by extrusion of a polycarbonate melt to the polycarbonate layer of a plastics composite of polycarbonate and fluoropolymer. In this case, the layer of polycarbonate originally present and the additionally applied layer of polycarbonate combine to form a single polycarbonate layer, so that a plastics composite of two layers, a fluoropolymer layer and a polycarbonate layer, is obtained. The plastics composite of polycarbonate and fluoropolymer used, onto which the polycarbonate melt is applied, may contain a coupling agent layer between the fluoropolymer layer and the polycarbonate layer. Suitable coupling agents in this case are, for example, acrylate polymers.

A further preferred embodiment of the process according to the invention is that in which the two-layer plastics composite in process step a) is applied by extrusion of a polycarbonate melt to a solid fluoropolymer layer. A coupling agent layer may be used between the layer of polycarbonate and the layer of fluoropolymer.

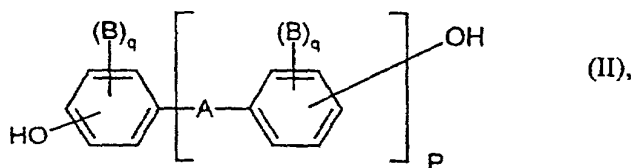
A further preferred embodiment of the process according to the invention is that in which the two-layer plastics composite in process step a) is prepared by bonding a layer of polycarbonate and a layer of fluoropolymer.

A particularly preferred embodiment of the process according to the invention is that in which the plastics composite from step a) has a temperature in process step b) at which

the ethylene-vinyl acetate copolymer does not undergo any appreciable thermal damage and at which the layer of ethylene-vinyl acetate copolymer is applied as a melt to the two-layer plastics composite. In this case, the process steps a) and b) are carried out preferably one immediately after the other, temperature control of the two-layer plastics composite taking place by cooling on a transport section.

Ethylene-vinyl acetate copolymers within the meaning of the present invention are copolymers of vinyl acetate and ethylene, which preferably have a vinyl acetate content of at least 30 wt.%. Particularly preferably, they have a vinyl acetate content from 50 to 90 wt.%. Such ethylene-vinyl acetate copolymers are well known and are sold as commercial products. For example, ethylene-vinyl acetate film, Elvax 485 type from BP Chemicals, D-89165 Dietenheim, Germany. They may be prepared by known methods.

Polycarbonates within the meaning of the present invention are those based on diphenols corresponding to the formula (II)



wherein

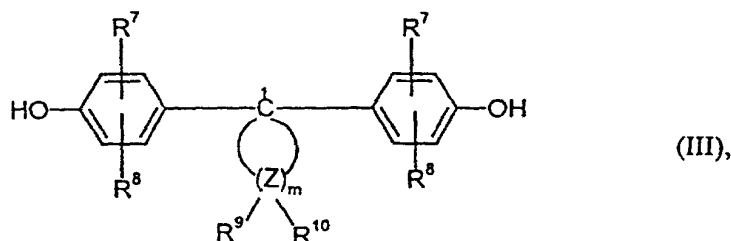
A means a single bond C<sub>1</sub>-C<sub>5</sub>-alkylene, C<sub>2</sub>-C<sub>5</sub>-alkylidene, C<sub>5</sub>-C<sub>6</sub>-cycloalkylidene, S or SO<sub>2</sub>,

B means chlorine, bromine, CH<sub>3</sub>,

q means 0, 1 or 2, and

p is 1 or 0,

or alkyl-substituted dihydroxyphenyl cycloalkanes corresponding to the formula (III)



wherein

$R^7$  and  $R^8$  independently of one another, mean in each case hydrogen, halogen, preferably chlorine or bromine,  $C_1$ - $C_8$ -alkyl,  $C_5$ - $C_6$ -cycloalkyl,  $C_6$ - $C_{10}$ -aryl, preferably phenyl, and  $C_7$ - $C_{12}$ -aralkyl, preferably phenyl- $C_1$ - $C_4$ -alkyl, particularly benzyl,

$m$  means an integer of 4, 5, 6 or 7, preferably 4 or 5,

$R^9$  and  $R^{10}$  which may be selected individually for each Z, independently of one another mean hydrogen or  $C_1$ - $C_6$ -alkyl,

and

Z means carbon, provided that on at least one atom Z  $R^9$  and  $R^{10}$  simultaneously mean alkyl.

Suitable diphenols corresponding to the formula (II) are, e.g., hydroquinone, resorcinol, 4,4'-dihydroxydiphenyl, 2,2-bis-(4-hydroxyphenyl) propane (i.e. bisphenol A), 2,4-bis-(4-hydroxyphenyl)-2-methylbutane, 1,1-bis-(4-hydroxyphenyl)cyclohexane, 2,2-bis-(3-chloro-4-hydroxyphenyl)propane, 2,2-bis-(3,5-dibromo-4-hydroxyphenyl)propane.

Preferred diphenols corresponding to the formula (II) are 2,2-bis-(4-hydroxyphenyl)propane, 2,2-bis-(3,5-dichloro-4-hydroxyphenyl)propane and 1,1-bis-(4-hydroxyphenyl)cyclohexane.

Preferred diphenols corresponding to the formula (III) are 1,1-bis-(4-hydroxyphenyl)-3,3-dimethylcyclohexane, 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane and 1,1-bis-(4-hydroxyphenyl)-2,4,4-trimethylcyclopentane.

5 Polycarbonates suitable according to the invention are both homopolycarbonates and copolycarbonates. A mixture of the thermoplastic polycarbonates defined above is also suitable.

10 Polycarbonates may be prepared in a known way from diphenols with phosgene by the phase boundary process or with phosgene by the process in the homogenous phase, the so-called pyridine process, or by the melt transesterification process from diphenols and carbonic acid esters, wherein the molecular weight may be adjusted in a known way by an appropriate amount of known chain terminators. Said methods of preparation are described, e.g., in H. Schnell "Chemistry and Physics of Polycarbonates", Polymer Reviews, Volume 9, p. 31-76 Interscience Publishers, 1964.

15 Suitable chain terminators are, e.g., phenol, cumylphenol, p-chlorophenol, p-tert-butylphenol or 2,4,6-tribromophenol, or long-chain alkylphenols, such as 4-(1,1,3,3-tetramethylbutyl)phenol or monoalkylphenol, or dialkylphenol with a total of 8 to 20 carbon atoms in the alkyl substituents such as, e.g., 3,5-di-tert.-butylphenol, p-isooctylphenol, p-tert.-octylphenol, p-dodecylphenol and 2-(3,5-dimethylheptyl)phenol and 4-(3,5-dimethylheptyl)phenol.

20 The amount of chain terminators is generally from 0.5 to 10 mole %, based on the sum of the diphenols used in each case corresponding to the formulae (II) and/or (III).

The polycarbonates suitable according to the invention have average molecular weights ( $\bar{M}_w$ , weight-average, measured, e.g., by ultracentrifugation or scattered light measurement) from 10,000 to 200,000 g/mole, preferably 18,000 to 80,000 g/mole, particularly preferably 19,000 to 38,000 g/mole.

25



1,1,1-tris(4-hydroxyphenyl)ethane and bis-(3-methyl-4-hydroxyphenyl)-2-oxo-2,3-dihydroindole is preferred.

The polycarbonates suitable according to the invention may be branched in a known manner, and preferably by the incorporation of 0.05 to 2 mole %, based on the sum of  
5 diphenols used, of compounds with three or more than three functional groups, e.g., those with three or more than three phenolic groups.

Preferred polycarbonates apart from the bisphenol-A homopolycarbonate, are the copolycarbonates of bisphenol A with up to 15 mole %, based on the mole sums of  
10 diphenols, of 2,2-bis-(3,5-dibromo-4-hydroxyphenyl)propane and the copolycarbonates of bisphenol A with up to 60 mole %, based on the mole sums of diphenols, of 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane.

The polycarbonates may be replaced partially or wholly by aromatic polyester carbonates. The aromatic polycarbonates may also contain polysiloxane blocks. The preparation thereof is described, for example, in US-A 3 821 325.

15 The fluoropolymers used are polymers in which the hydrogen atoms of the carbon chain of polyethylene are replaced wholly or partially by fluorine atoms, and chlorine or fluorine-chlorine derivatives thereof and derived copolymers.

The fluoropolymers used in preference are polyvinyl fluoride or polyvinylidene fluoride. Polyvinyl fluoride is particularly preferred.

20 A plastics composite within the meaning of the present invention is, in particular, a plastics sheet or a plastics film. The thickness of the sheet or film is preferably 300  $\mu\text{m}$  to 12 mm, particularly preferably 500  $\mu\text{m}$  to 5 mm. The thickness of the fluoropolymer layer is preferably 10  $\mu\text{m}$  to 500  $\mu\text{m}$ , particularly preferably 20  $\mu\text{m}$  to 200  $\mu\text{m}$ . The thickness of the polycarbonate layer is preferably 100  $\mu\text{m}$  to 12 mm, particularly

preferably 200  $\mu\text{m}$  to 5 mm. The thickness of the ethylene-vinyl acetate copolymer layer is preferably 100  $\mu\text{m}$  to 1 mm, particularly preferably 300  $\mu\text{m}$  to 800  $\mu\text{m}$ .

The plastics composites according to the invention may be used, for example, for the manufacture of photovoltaic modules. For example, solar cells based on silicon may be embedded between two plastics composites with the three-layer composition fluoropolymer-polycarbonate-ethylene-vinyl acetate copolymer in such a way that the sequence of layers is fluoropolymer-polycarbonate-ethylene-vinyl acetate copolymer-solar cells-ethylene-vinyl acetate copolymer-polycarbonate-fluoropolymer. In a subsequent process, e.g., the hot laminating process, this arrangement is heated to such an extent that the EVA softens so that the two EVA layers combine with the inclusion of the solar cells.

The plastics composites according to the invention may be used for numerous other purposes. For example, due to the good resistance to chemicals of the fluoropolymer layer and due to their good resistance to weathering and UV resistance, they may be used, optionally after being laminated onto support materials by means of the ethylene-vinyl acetate copolymer layer, as a material in the chemical industry, the food industry or the pharmaceutical industry.

The process according to the invention is explained in more detail below on the basis of a drawing (Fig. 1) representing only one preferred embodiment.

The preparation of a plastics composite of three layers according to the invention is shown in Fig. 1. A two-layer film 1 of a polycarbonate layer 2 and a polyvinyl fluoride layer 3 is guided over a three-roll polishing stack 4. A melt film of polycarbonate is fed between the first and the second roll of the polishing stack through a slot die 5 which is fed with a polycarbonate melt by means of an extruder. Said melt film combines with the polycarbonate layer 2 so that a thicker, still hot polycarbonate layer and hence in turn a two-layer film 6 is obtained. This is cooled by transport on a roller conveyor 7 to

the required temperature at which the film of ethylene-vinyl acetate copolymer 9 is no longer damaged by heat but still softens sufficiently to bring about good adhesion. A film of ethylene-vinyl acetate copolymer 9 is then applied to the polycarbonate layer. The plastics composite of three layers 10 according to the invention obtained in this way is hauled off over two further rollers 11 and 12 and may then be wound, optionally after cooling, or further processed in another way.

The invention will be explained subsequently in more detail on the basis of examples.

With an arrangement as shown in Fig. 1, an EVA film was applied during PC sheet extrusion to the upper side of the sheet, and a PVF-PC film to the lower side of the sheet.

The PC sheet was prepared by extrusion through a slot die with a width of 800 mm and a die lip gap of 5 mm. The PC sheet had a thickness of 3 mm and a width of 780 mm.

The polycarbonate used was Makrolon® 3103 from Bayer AG, Leverkusen, Germany. This is a homopolycarbonate based on bisphenol A.

The processing parameters typical for Makrolon® 3103 sheet extrusion were set:

**Temperature settings on the extruder:**

Extruder zone Z1 290°C

Extruder zone Z2 290°C

Extruder zone Z3 280°C

Extruder zone Z4 260°C

Extruder zone Z5 265°C

Extruder zone Z6 290°C

Die 280°C

Mass temperature 300°C

**Nature of the film:**

5

1. Ethylene-vinyl acetate copolymer (EVA): Elvax® 485.00, BP Chemicals D-89165 Dietenheim, Germany

Film thickness: 500 µm

Film width: 660 mm

10

2. Two-layer film 1 (Makrofol® EPC, (PVC-PC film), Bayer)

Film thickness 550 µm (375 µm bisphenol A homopolycarbonate, 25 µm PVF)

15

The EVA film was not dried beforehand, the Makrofol EPC film was pre-dried at 80°C in the circulating air drier.

Roll temperatures of the three-roll polishing stack:

Roll 1: 120°C

Roll 2: 127°C

Roll 3: 143°C

20

Haul-off speed: 0.98 m/min

**Sheet samples prepared:**

1. PVF-PC sheet
2. PVF-PC-EVA sheet

**Test procedure:**

- 5 The PVF-PC film was fed from below over the 1st roll to the 1st nip. The PC side of the film (smooth side of the film) was in contact with the PC melt. The bead remained on roll 2 (contrary to the experience with material laminating). The film was fed via the traverse, the film being kept somewhat under tension. The unwinding speed was established by the sheet haul-off speed.
- 10 The EVA film was fed from above over a chromium roller (11), the sheet being passed through beneath the roller (film feed from above, pressure applied by the roller). The sheet temperature at this location was 76°C. In this position, the film started to melt easily and could not be separated again by hand. The unwinding speed was established by the sheet haul-off speed.
- 15 In order to prevent the EVA film sticking to the chromium rollers of the roller conveyor, a protective film of PE was fed between EVA film and chromium roller, which acted simultaneously as a protective film for the composite. The PE film could be removed later from the laminated sheet without difficulty.

**Patent Claims**

1. A process for the preparation of plastics composites of three layers, wherein the first layer comprises a fluoropolymer, the second, middle layer comprises polycarbonate and the third layer comprises ethylene-vinyl acetate copolymer, characterised by the following steps to be executed one after the other:
  - a) preparation of a plastics composite of two layers, wherein the first layer comprises the fluoropolymer and the second layer comprises the polycarbonate,
  - b) application of the third layer of ethylene-vinyl acetate copolymer to the polycarbonate layer of the plastics composite of two layers.
2. A process according to claim 1, characterised in that the plastics composite of two layers in the process step b) has a temperature at which the ethylene-vinyl acetate copolymer does not undergo thermal damage, and in that the layer of ethylene-vinyl acetate copolymer is applied as a film to the plastics composite of two layers.
3. A process according to claim 2, characterised in that the plastics composite of two layers in process step b) has a temperature from 20°C to 120°C.
4. A process according to one of claims 1 to 3, characterised in that the process steps a) and b) are carried out one immediately after the other.
5. A process according to one of claims 1 to 4, characterised in that the fluoropolymer used is polyvinyl fluoride and the polycarbonate used is polycarbonate based on bisphenol A.







## COMBINED DECLARATION AND POWER OF ATTORNEY

ATTORNEY DOCKET NO

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought

on the invention entitled

**MULTILAYER PLASTIC COMPOSITES AND A METHOD FOR THE PRODUCTION THEREOF**

the specification of which is attached hereto,

or was filed on **August 18, 2000**

as a PCT Application Serial No. **PCT/EP00/08070**

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s), the priority(ies) of which is/are to be claimed:

**199 41 214.6**  
(Number)

**Germany**  
(Country)

**August 30, 1999**  
(Month/Day/Year Filed)

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose the material information as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)

(Filing Date)

(Status)

(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)

(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Le A 33 536-US

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

JOSEPH C. GIL, Patent Office Registration Number 26,602; ARON PREIS, Patent Office Registration Number 29,426; LYNDANNE M. WHALEN, Patent Office Registration Number 29,457; THOMAS W. ROY, Patent Office Registration Number 29,582; RICHARD E. L. HENDERSON, Patent Office Registration Number 31,619; GODFRIED R. AKORLI, Patent Office Registration Number 28,779; N. DENISE BROWN, Patent Office Registration Number 36,097; NOLAND J. CHEUNG, Patent Office Registration Number 39,138; DIDERICO VAN EYL, Patent Office Registration Number 38,641; CAROLYN M. SLOANE, Patent Office Registration Number 44,339; JAMES R. FRANKS, Patent Office Registration Number 42,552; JACKIE ANN ZURCHER, Patent Office Registration Number 42,251; RAYMOND J. HARMUTH, Patent Office Registration Number 33,896; JOHN E. WROZINSKI, JR., Patent Office Registration Number 46,179; JENNIFER R. SENG, Patent Office Registration Number 48,851, all of Bayer Corporation, Pittsburgh, Pennsylvania 15205-9741

Send Correspondence To:  
Patent Department  
Bayer Corporation  
100 Bayer Road  
Pittsburgh, Pennsylvania 15205-9741

Direct Telephone Calls To:  
  
(412) 777-2349

FULL NAME OF SOLE OR FIRST INVENTOR Jürgen Röhner		INVENTOR'S SIGNATURE <i>J. Röhner</i>	DATE 16.01.02
RESIDENCE D 51069 Köln, Germany DEX		CITIZENSHIP German	
POST OFFICE ADDRESS c/o BAYER AKTIENGESELLSCHAFT, D 51368 Leverkusen, Germany			
FULL NAME OF SECOND INVENTOR Klaus Zander		INVENTOR'S SIGNATURE <i>Klaus Zander</i>	DATE 18.01.02
RESIDENCE D 45478 Mülheim, Germany DEX		CITIZENSHIP German	
POST OFFICE ADDRESS c/o BAYER AKTIENGESELLSCHAFT, D 51368 Leverkusen, Germany			
FULL NAME OF THIRD INVENTOR Rüdiger Gorny		INVENTOR'S SIGNATURE <i>Rüdiger Gorny</i>	DATE 2002-01-21
RESIDENCE D 47800 Krefeld, Germany DEX		CITIZENSHIP German	
POST OFFICE ADDRESS c/o BAYER AKTIENGESELLSCHAFT, D 51368 Leverkusen, Germany			
FULL NAME OF FOURTH INVENTOR Marco Roelofs		INVENTOR'S SIGNATURE <i>Marco Roelofs</i>	DATE 21. Jan. 2002
RESIDENCE D 47807 Krefeld, Germany DEX		CITIZENSHIP German	
POST OFFICE ADDRESS c/o BAYER AKTIENGESELLSCHAFT, D 51368 Leverkusen, Germany			
FULL NAME OF FIFTH INVENTOR		INVENTOR'S SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF SIXTH INVENTOR		INVENTOR'S SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF SEVENTH INVENTOR		INVENTOR'S SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			

Le A 33 536-US